Role and behavior of the orbital moment in magnetic phase transitions

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INTRODUCTION

In the **RCo_2** "Laves-phases" compounds (where R is a lanthanide ion) the electronic structure of the d band is near the critical condition for the Co-magnetic moment formation. In comparison, Ni and Fe Laves-phases are well below and above the condition of moment formation, respectively. This peculiar situation renders the magnetism of the Co Laves phases very rich and interesting.

When formed with non-magnetic R elements (or Y), **RCo_2** are exchange-enhanced paramagnets. The Co moment can be induced in those compounds by applying very high magnetic fields (70 T), where they exhibit a metamagnetic transition to a ferromagnetic state.

In contrast, in the RCo_2 compounds with a magnetic rare-earth, the internal f-d exchange field polarizes the Co sub bands, driving the magnetic moment formation. As in every 3d-4f intermetallic, the intersublattice exchange and the 3rd Hund's rule turns the material into a ferromagnet for light rare earths or a ferrimagnet for heavy rare earths[1].

MOTIVATIONS

During our scheduled beamtime at ALS in 2002, we did study **$ErCo_2$**. Er is a heavy rare earth, so $ErCo_2$ is a ferrimagnet with Curie temperature T_c = 35 K. The Er sublattice is always dominant, as the intrinsic Er moment is large compared to the induced Co moment. When a magnetic field is applied, the Er moment is parallel to the field at all temperatures.

Another interesting effect observed in the RCo_2 compounds is that with R = Dy, Ho and Er the magnetic phase transition at T_C is of a first-order type. This phenomenon is however intimately related with the metamagnetic properties of the d subsystem[2]: Co suffers a metamagnetic transition at T_c in which a magnetic moment of about 1 μ_B per Co atom is developed below T_c (Co moment show a clear discontinuity) giving rise to a first order magnetic transition at T_c. The Co magnetism in this material is a consequence of the itinerant electron metamagnetism induced in a large exchange field appearing at T_C where the localized Er moments order ferromagnetically. Indeed T_c shifts to higher values with increasing the applied field (T_c = 32K at 0.1T and T_c = 42K at 5T). [2]

1st order Curie points are only possible if there are magnetovolumic effects associated with the ordering transition. Givord[3] corroborated this in 1972 by measuring the lattice parameters as a function of temperature. Gratz et al. Have determined that below Tc, the cubic unit cell of RCo_2 is distorted due to the appearance of a spontaneous anisotropic magnetostriction, attributed to the existence of an orbital part in total M_{Co}. Our main goal in this study was to measure the orbital moment behaviour through the phase transition, which must be at the origin of this spontaneous anisotropic magnetostriction.

EXPERIMENTS AND RESULTS

In order to study the Co orbital magnetic moment we have measured XMCD spectra at the Co $L_{2,3}$ and Er

$M_{4,5}$ edges as a function of temperature and applied field on an $ErCo_2$ ingot. The experiments have been performed at BL4.0.2, using the 6T-2K XMCD endstation from the University of California at Davis and Berkeley Lab (Dr. P. Cramer, spokesperson). In Figure 1 we show typical spectra obtained:

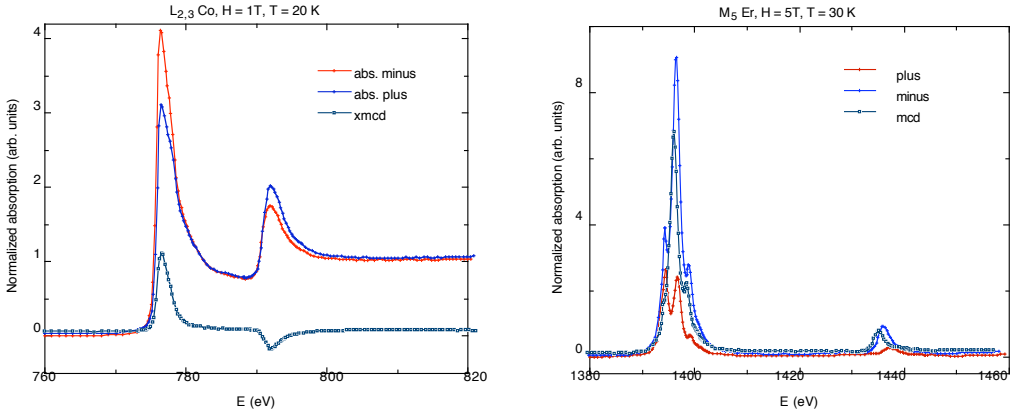

Fig. 1. Co and Er absorption edges and magnetic dichroism in the ferrimagnetic region.

Er MCD signal as a function of temperature and magnetic field follows in first approximation the magnetization bevavior (measured with SQUID magnetometry), indicating that the Er net magnetic moment is alligned parallel to the applied field, both in the paramagnetic and the ferrimagnetic phase.

Co net magnetic moment is polarized parallelly to the applied field (and the Er moment) at high temperatures but orders antiparallel below T_c, as expected for a ferrimagnet: we clearly observe the temperature, T_f, at which the Co changes its orientation relative to the field, ($T_f > T_c$). Indeed, one can argue that Co moment orientation will change when the local exchange field created by the Er sub-lattice is higher than the external applied one.

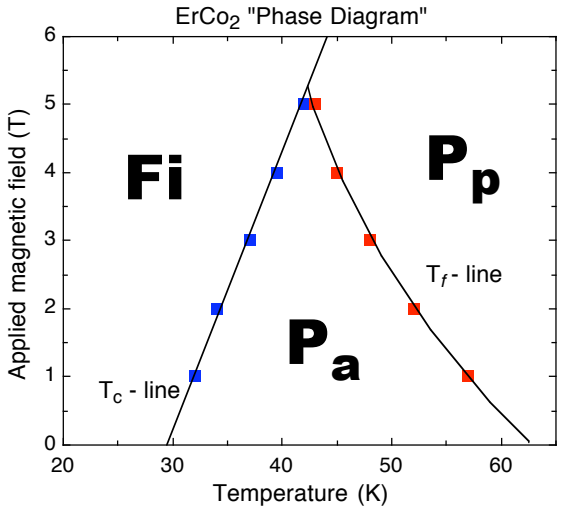

Fig. 2. Phase diagram of ErCo2 as obtained from our XMCD data. The ferrimagnetic (Fi) is separated by a "true" 1st order phase-transition line while the paramagnetic phase is divided into parallel and antiparallel "phases" by a flipping temperature line defined by the zero-Co XMCD points (red squares).

In agreement with this argument we find experimentally that the temperature difference between T_c and T_f decreases with increasing H (22K at 1T, but only 2K at 5T).

By applying the well known "sum-rules" to our XMCD data we can obtain the orbital and spin moment per Co-3d hole. A preliminary analysis is shown on figure 3. Left panel shows the absolute value of the

orbital moment *per hole* obtained from our XMCD data below T_f, which shows a clear jump below the Curie point. In a different scale the total magnetization is shown for comparison.

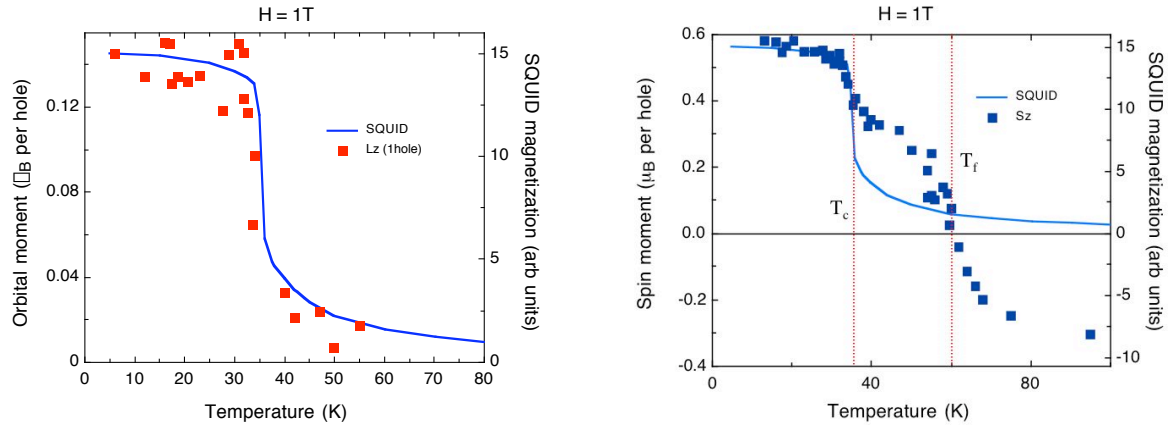

Fig.3. Orbital and spin moments of Co compared with total magnetization.

Right panel of Figure 3 shows Right panel of Figure 3 the spin moment per 3d-hole as a function of temperature. It has to be understood that the actual moments above T_c are much smaller than in the ordered region, as the internal field is not able to "develop holes" in the 3d band, and consequently the moment "per hole" is affected by a much smaller *"nr. of holes"* factor above T_c. This causes the apparent departure of the spin moment data from the macroscopic magnetization. Note also that, for the sake of clarity, the spin moments are inverted from their actual orientation. Below T_c, the moment per hole is in good agreement with the occupation numbers and magnetic moments in literature (see the cited Gratz's review).

Our project is continuing at ALS, and will include measurements of **$NdCo_2$** (a 2nd order transition ferromagnet) and **$TbCo_2$** (a 2nd order transition ferrimagnet). With the study of these samples we hope to be able to clarify the orbital effects induced in the Co moments by short-range as well as 1st and 2nd order long-range magnetic transitions.

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The research interests of the group, a summary of results and a publication list are presented on the World Wide Web at http://icma.csic.unizar.es/nivel2/lin_inv/5_leis/chaboy.htm